# Caloric curve in fragmentation

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## Abstract

We study phase transitions of small two dimensional Lennard Jones drops via microcanonical molecular dynamics in a broad energy range. We found that the caloric curve can be extended to high energies to comprehend high evaporation rates and highly non-equilibrium phenomena such as multifragmentation. Multifragmentation appears as a constant temperature region in the caloric curve like the solid-liquid phase transition.

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The problem of phase transitions in small many body systems has attracted much attention in different areas of physics during the last two decades and remains of great importance. Among these processes, and particularly important in nuclear physics we can mention multifragmentation. In this phenomena a very excited small system undergoes a highly non equilibrium process, in which a collective expansion develops and the initial drop breaks up in several intermediate mass fragments. These fragmentation processes are studied in many branches of physics, for example fragmentation in intermediate mass heavy ion collisions [5], drop formation in fluids flowing through nozzles, lithium jets bombarded with neutrons in inertial confinement reactors [6], fragmentation in cluster deposition for the synthesis of nanostructured surfaces [7]. In spite of the great efforts done is order to study these phenomena many questions remain unsolved, the mechanisms that make the excited drop break into several fragments and the possible connection of these processes with the liquid-vapor phase transition are not clear at present [8]. In the nuclear multifragmentation case, we would like to mention the current efforts towards the determination of the caloric curve by the ALADIN collaboration [9], and the calculations of different critical exponents by the EOS collaboration [10].

When dealing with infinite systems one usually thinks on solid-liquid and liquid-vapor phase transitions. It is well known that small finite systems display solid-like to liquid-like transition, which can be thought of as the rounding-off of the first order phase transition of the infinite case due to finite size effects. On the other hand the process of fragmentation described above is a finite system thing, and does not appear in the thermodynamic limit.

The analysis of the solid-like to liquid-like phase transition of very small systems, and its connection to the macroscopic solid-liquid phase transition has relied strongly on computer simulations. One of the most relevant quantities regarding this phenomena is the caloric curve $(T(E))$, which displays a rather constant region at the phase transition. As expected for small systems the behavior depends on the thermodynamic ensemble used in the simulation [1]. Different simulation methods were used, for example constant temperature molecular dynamics [2], constant $(T, P, N)$ Monte Carlo [3] and microcanonical molecular

dynamics [4]. The upper limit of the range of energies studied in these works is chosen so as to avoid dealing with evaporation. Although this phase transition is not relevant for the nuclear case, we will show that its study can be of great importance in order to understand fragmentation in classical many body systems.

Consider a small isolated drop in an infinite volume; according to the energy of the drop we classify schematically its behavior in three different cases. a) Low energy drops, where evaporation does not occur, at least during the time scales considered, this case contains the solid-like to liquid-like phase transition. For higher energy we will get b) evaporating drops, where few particles evaporate during the experiment. It is worth mentioning at this point that this kind of experiment, as any of this kind with free boundary conditions, is to be thought as equivalent to a time-scale constraint in the experiment conducted at zero pressure [3]. For even higher energy c) we enter the case of multifragmentation. The initial state of this experiment is a hot and compressed drop which expands and breaks up in several fragments. It was shown in [11] that some of the initial thermal excitation is transformed, via inter-particle collisions, into a collective radial expansion, and consequently the system cools down. The time scale considered in the present work is related to the one of fragment formation for the multifragmentation experiments [11].

In this communication we study the behavior of two dimensional drops in a wide energy range, embodying the solid-like to liquid-like transition, evaporating drops, and the phenomena of fragmentation. We first describe the microscopic model that we use to simulate the atomistic system. We then present our multifragmentation computer experiments and the information that can be extracted from them, followed by our results for equilibrated drops, which contains the solid-liquid phase transition. The connection of the two phenomena is then shown.

We simulate the atomistic two dimensional system formed by $N = 100$ Lennard Jones particles interacting via the two body potential:

$$V(r) = 4\epsilon \left[ \left( \frac{\sigma}{r} \right)^{12} - \left( \frac{\sigma}{r} \right)^{6} - \left( \frac{\sigma}{r_c} \right)^{12} + \left( \frac{\sigma}{r_c} \right)^{6} \right], \tag{1}$$

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where $r_c$ is the cutoff radius, the potential is taken as zero for $r \geq r_c$. We took $r_c = 3\sigma$. The unit of time and energy are $t_0 = \sqrt{\sigma^2 m / 48\epsilon}$ and $\epsilon$ respectively. The time step that we used for the integration of the equation of motion was fixed to $t_{int} = 0.0025 t_0$, in this way the energy and total angular momentum is conserved better than 0.01%.

For the study of multifragmentation we analyzed the time evolution of compressed and hot drops. The initial configurations are obtained by cutting a circular drop from a thermalized two dimensional Lennard Jones periodic system. These initial configurations are macroscopically characterized by the energy $E$, the density $\rho$ (of the periodic system) and the number of particles which is fixed to $N = 100$, see [11] for details.

Defining the temperature in these experiments is not an easy task. The process of fragmentation can be divided in two different stages. During the first stage of the evolution "flux and fragment formation stage" a collective expansion builds up, and the asymptotic clusters are formed in phase space, according to the ECFM (early cluster formation model) [12,11]. During the second stage "fragment emission stage" the clusters, already formed in phase space [11], spread in coordinate space, i.e. they are emitted. It is worth mentioning at this point that the ECFM allows us to preciselly identify the break-up or fragment formation time.

In order to study the cooling of the system while the expansion builds up, we can define for the initial dense stages a local temperature, which depends on time and position, in the following way. We define the local temperature $T_l(\mathbf{r}, t)$ as the velocity fluctuations around the mean radial expansion, which is position dependent (the outer regions expand faster than the inner ones). In other words we make the conjecture that our system is in local equilibrium [13], i.e. the velocity distribution of all the particles of our system depend on position and satisfy:

$$f(\mathbf{v}; \mathbf{r}) = \rho(\mathbf{r}) \left( \frac{m\beta(\mathbf{r})}{2\pi} \right) e^{\beta(\mathbf{r})\frac{m}{2}(\mathbf{v} - \mathbf{v}_{rad}(\mathbf{r}))^2} \tag{2}$$

where $\rho(\mathbf{r})$ and $\beta(\mathbf{r})$ are the density and inverse of the temperature respectively; $\mathbf{v}_{rad}(\mathbf{r})$ is the collective velocity which in our case, due to the symmetry of the problem, is in the radial

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direction. The actual calculation is done in the following way: we divide the system in ten concentric circular regions, centered in the c.m. of the system, and we calculate the mean radial velocity of each circular ring. The temperature of each circular region is defined as the velocity fluctuations around the meassured collective motion (mean radial velocity) of the corresponding ring, [11]. The total kinetic energy is the sum of the one corresponding to the expansion and that associated to the local temperature. The general picture obtained is the following: the process involves a fast cooling while the expansion builds up, and after a time $\tau(E)$ the temperature remains almost constant in time [11]. Furthermore the temperature of the central regions reach approximately the same value and the radial and transverse velocity fluctuations (or local temperatures) are very similar. This last fact shows that the system reaches some degree of local thermalization [11], in this way we see that our conjecture of local equilibrium is justified.

We can then define the temperature of the system as a whole, $\langle T_l(t) \rangle$, as the average over the temperatures of the inner regions so as to include approximately 80 particles; the outer rings are disregarded because they are populated with only few free particles and very small aggregates. This definition is only valid as long as the system is still dense.

In Fig. 1 we show $\langle T_l \rangle$ as a function of time, for different initial energies and densities, see caption for details. It can be seen the fast cooling during the first stage of the evolution. The initial conditions shown in Fig. 1 represent a very wide range of energies as reffers to the fragmentation pattern (the initial temperature range is $1\epsilon \leq \langle T_l(0) \rangle \leq 4\epsilon$). For $E = 0.3\epsilon$ the system breaks into several small fragments and the mass spectrum is exponentially decaying, while for $E = -1.1\epsilon$ the mass spectrum is U shaped, at this low energy an important part of the system remains bound, see [11]. From Fig.1 we get the important result that while the collective expansion builds up the system cools down until the local temperature reaches a given value, which lies between $0.35\epsilon$ and $0.4\epsilon$, independent of the initial conditions, i.e. density and energy. From this time on, the system continues its expansion at a constant radial velocity and breaks-up.

Asymptotically the system is formed by a set of non interacting clusters that are moving

away from each other, it is clear then that we can not speak of the temperature of the system as a whole. In this case, when fragments are already formed, the total kinetic energy of the system can be expressed in the following way

$$K_{total} = \sum_{clusters} \frac{1}{2m_c} \mathbf{P}_c^2 + \sum_{clusters} K_i^{c.m.} + \sum_{clusters} \frac{I_c \omega_c^2}{2} \tag{3}$$

where $P_c$ is the momentum of the center o mass of cluster $c$ of total mass $m_c$ and the last term is the total rotational kinetic energy of the clusters. $K_i^{c.m.}$ is, then, the kinetic energy of particles belonging to cluster $c$ once the total linear and angular momentum are removed.

We now focus on the temperature of the asymptotic fragments $T_f$, which is given by :

$$T_f(i) = \frac{1}{n_i - 3/2} K_i^{c.m.}, \tag{4}$$

where $n_i$ denotes the mass number of cluster $i$. In Fig. 2 we show the average temperature of the asymptotic fragments of our molecular dynamics experiments as a function of their mass, for four different initial conditions: $E = -1.1\epsilon$ , $\rho = 0.75\sigma^{-2}$ (full circles), $E = -0.55\epsilon$, $\rho = 0.75\sigma^{-2}$ (empty circles), $E = 0.8\epsilon$, $\rho = 0.75\sigma^{-2}$ (diamonds) $E = -0.3\epsilon$ , $\rho = 0.85\sigma^{-2}$ (triangles). No less than 100 events for each initial condition were averaged. Again the cases shown in Fig. 2 represent a wide range of excitation as refers to the behavior of the system. It can be seen that for $E = 0.8\epsilon$ there are no clusters of mass bigger that 20, at this high energy the system breaks in several small mass fragments. On the other hand, for $E = -1.1\epsilon$, there are very few intermediate mass fragments which comes from a U shaped mass spectra, the mass spectra for most of the cases are shown in [11]. Fig. 2 contains a remarkable result: despite the wide range of excitations shown, the temperature of clusters with a given mass number $n$ is fixed no matter the initial condition of the system.

It is important to note that the temperature of the intermediate and big mass clusters is equal to the local temperature (velocity fluctuations around the mean radial velocity as defined above) at the break up time (see Fig. 1). In order to gain a better comprehension of the multifragmentation process, it is important to understand what is so particular about this value of temperature, so we studied our $N = 100$ LJ particles system for lower energies, when fragmentation does not occur.

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The study of the low energy regimes of small systems usually involve the calculation of caloric curves $(T(E))$, which might give us some information about the meaning of the value of temperature discussed in the last paragraph. The usual caloric curves show three regions: a) solid region in which $T$ increases steadily leading to constant specific heat, b) the solid-like to liquid-like transition in which $T$ is rather constant, c) liquid region in which, again, $T$ increases [1]. It is clear that the liquid-like state cannot be heated without limits, so another plateau is to be expected.

In order to study this we calculated the caloric curve of our two dimensional Lennard Jones drops. We imposed free boundary conditions to our system. In this way we reproduce the conditions at which multifragmentation experiments take place, where the system is not confined. The initial configuration was constructed in the same way as for multifragmentation, but this time the energy and the density were close to those of the ground state of the drop. When the drop is evolved the particles display approximately a triangular lattice; the system does not explode, nor does it evaporate particles. In order to study the caloric curve $(T(E))$, after performing the isoenergetic evolution of the drop the velocities of the particles of the last configuration are re-scaled in order to increase the energy in a constant amount, and a new isoenergetic evolution is performed. This procedure is repeated in order to cover the desired range of energies. If the system, in its evolution, evaporates one or more particles they are disregarded for the next evolution, at a slightly higher energy; only the particles remaining in the drop are considered in the velocity re-scaling after their center of mass velocity and angular momentum are removed. With our procedure the system will loose particles, due to evaporation, as it is heated. For our calculations we only considered configuration in which the number of particles in the remaining drop was $90 \leq N \leq 100$. This variation in the mass of the system is not a serious problem since we found that the temperature and other macroscopic quantities of our system do not change when few particles are evaporated.

The temperature for a given energy is given by the well known relation for 2-D:

$$T = \left\langle \frac{1}{N(t) - 3/2} \sum_{i=1}^{N(t)} \frac{1}{2} m \mathbf{v}_i^2 \right\rangle_t , \tag{5}$$

where $\langle \rangle_t$ means time average and $N(t)$ is the number of particles which at time $t$ have not evaporated. The time average is done once the system has reached equilibrium, the first $2 \times 10^4$ time steps accomplish this thermalization. The total time considered for the average is $150 t_0$ which is approximately the time of the multifragmentation experiments. It is worth mentioning that this last definition of temperature is equivalent to those used in multifragmentation because for these low energy cases the collective expansion can be disregarded, no more than three particles are evaporated for a given energy in the period of time considered.

In this way we studied the range of energy $-2.8\epsilon$ to $-1.4\epsilon$ of the caloric curve, see the circles in Fig. 3. Three different regions can be identified. I) Solid region in which $T$ increases steadily, II) solid-liquid transition, in which $T$ is rather constant, and III) liquid region in which $T$ increases steadily again and then presents the beginning of a high energy plateau $(-1.8\epsilon \leq E \leq -1.4\epsilon)$. Although the plateau denoting the solid-like to liquid-like phase transition is very small, the transition as one increases the energy is also denoted by particles being popped out of the surface and occupying the adjacent outer layer [2] and by the sudden increase of the root mean square bond length fluctuation (Lindemann index) above 0.1 [4] (not shown in the figure for clarity). The solid-like to liquid-like phase transition in small clusters has been extensively studied, see references in the first paragraph, and although this is, to the best of our knowledge, the first time that this is done in two dimensional systems, we are not interested in a detailed description of the solid-liquid phase transition but in the general behavior of the caloric curve and its connection with multifragmentation.

In the high energy part of region III the evaporation is quite important and it is very difficult to attain metastable liquid-like configurations for long enough times if the energy is increased beyond $-1.4\epsilon$, this fact makes the calculation of the caloric curve by this procedure meaningless beyond this energy. This is because for higher energies we enter the multifragmentation regime, described above. Remember that in this case the initial state of

the system is out of equilibrium and characterized by energy and the density and the main de-excitation channel is the creation of a collective expansion, the system does not simply evaporate particles but expands and breaks into several fragments. The values of $T$ for energies higher that $-1.4\epsilon$ in Fig. 3 were obtained from the results shown in figures 1 and 2. The temperature corresponding to a given energy is calculated in two ways. a) squares show the local temperature at break-up time, $\langle T_l \rangle$ averaged over a period of time of approximately $30t_0$ centered at the break up time (the break-up times can be found in [11]). This is basically the constant value of the local temperature shown in Fig. 2. b) The diamonds show the average cluster temperature (we only considered fragments of mass number greater that 15). See caption for the initial densities. It can be seen that the asymptotic temperature of the clusters is quite similar to the local temperature at the break-up time, moreover we get the important result that the low energy part of the caloric curve joins smoothly the results of the multifragmentation experiments. The full caloric curve features two plateaux, the low energy one is related to the solid-like to liquid-like transition, and the high energy ones denotes the evaporating liquid and multifragmentation. This behavior of the caloric curve is in agreement with very recent experimental results for the nuclear caloric curve [14,15]. On the other hand the experimental result of [9] show a caloric curve featuring a plateau followed by a rise of temperature for high excitation energy, which was denoted "gas phase". This "gas phase" behavior was also found in computer simulations of nuclear fragmentation by Bondorf et. al. [16]. In this last work the temperature is related to the c.m. velocity of the asymptotic clusters, making strong assumptions regarding the flow velocity distribution. Let us stress that our approach makes no assumptions about the radial flow, in this way we make sure that the collective motion is correctly calculated and subtracted in order to compute the temperature of the expanding system. We do not find any increase of temperature in the fragmentation region, and within our picture of fragmentation, explained in the next paragraph, no "gas phase" is to expected in fragmentation experiments. Its presence in [16] might be due to the fact that the collective motion is not taken into account properly.

From the results presented in Fig. 3, the connection between multifragmentation and

the liquid-gas phase transition appears. From the region of highly evaporating liquid of Fig. 3 it can be seen that it is not possible to increase the temperature of a liquid drop beyond some limit value which for our system is $\sim 0.37\epsilon$, we will call it the limit temperature; as we increase the energy, the evaporation will become more important but the system will not heat up. In our multifragmentation experiments the initial state is artificially constructed hotter than the limit case, in real systems this highly excited state is obtained via a sudden input of energy to the system, for example by collisions. As the system evolves it rapidly expands and cools down until the limit value for temperature is achieved and the system needs no further relaxation, only some evaporation will occur. If the energy of the initial condition is increased, the radial collective velocity will grow and the system will break into smaller fragments but the asymptotic temperature will not change, in the same way as in the solid-liquid phase transition in which an input of energy will not result in an increase of temperature but in melting. When the system breaks, the clusters will be as hot as they can, which is determined by the limit temperature, this is why the temperature of the asymptotic clusters as a function of their mass is independent of the initial conditions, (Fig. 2).

In conclusion we found that the microcanonical caloric curve can be extended to high energies in such a way that it can describe the non equilibrium process of multifragmentation, which appears as a constant temperature region. This shows that although fragmentation is not a phase transition in a strict sense, e.g. it does not show phase coexistence, it has some properties that are characteristic of phase transitions. One of these is the flat caloric curve, other being that the mass spectra is similar to those found in the liquid-gas phase transition (see [11]and references therein). We also found that during the first stages of multifragmentation the system transforms thermal motion into a more ordered collective expansion until it reaches the limit temperature, independent of the initial conditions, which is the maximum attainable temperature for a liquid drop. This fact enable us to know in advance the velocity fluctuations over the collective expansion when the fragmenting system breaks. This is the first step towards a full characterization of the break up state in multifragmentation, the other important ingredients being the radial velocity field and the

density fluctuations in phase space.

## ACKNOWLEDGMENTS

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FIGURES

FIG. 1. Local temperature as a function of time, for different initial conditions. Full line denote $\rho = 0.75(1/\sigma^2)$ and $E = -1.1\epsilon$, dotted line $\rho = 0.75(1/\sigma^2)$ and $E = -0.55\epsilon$, dashed line $\rho = 0.75(1/\sigma^2)$ and $E = 0.3\epsilon$ and dashed-dotted line $\rho = 0.85(1/\sigma^2)$ and $E = -0.3\epsilon$.

FIG. 2. Temperature of the asymptotic clusters as a function of the mass number, for different initial conditions. Full circles denote $\rho = 0.75(1/\sigma^2)$ and $E = -1.1\epsilon$, empty circles $\rho = 0.75(1/\sigma^2)$ and $E = -0.55\epsilon$, empty diamonds $\rho = 0.75(1/\sigma^2)$ and $E = 0.8\epsilon$ and empty triangles $\rho = 0.85(1/\sigma^2)$ and $E = -0.3\epsilon$.

FIG. 3. Caloric curve. Four regions can be identified: I solid-like, II solid-like to liquid-like phase transition, III liquid-like and IV multifragmentation. Circles denote results from the equilibrium experiments, squares denote time average of the local temperature centered at the break-up in fragmentation and diamonds show the mean temperature of asymptotic clusters of mass number $> 15$ in multifragmentation. The initial density in the multifragmentation experiments is $\rho = 0.75(1/\sigma^2)$ for all energies but for $E = -0.3\epsilon$ and $E = -0.5\epsilon$ in which it is $\rho = 0.85(1/\sigma^2)$
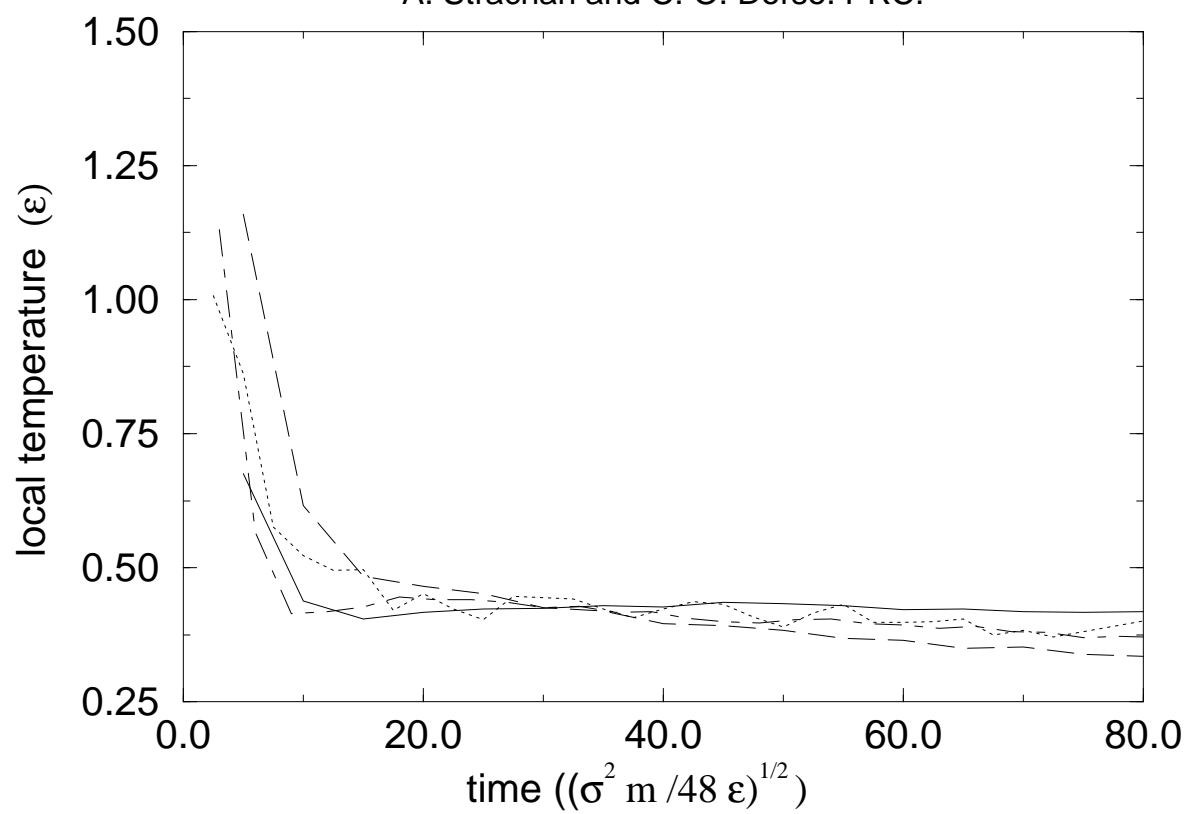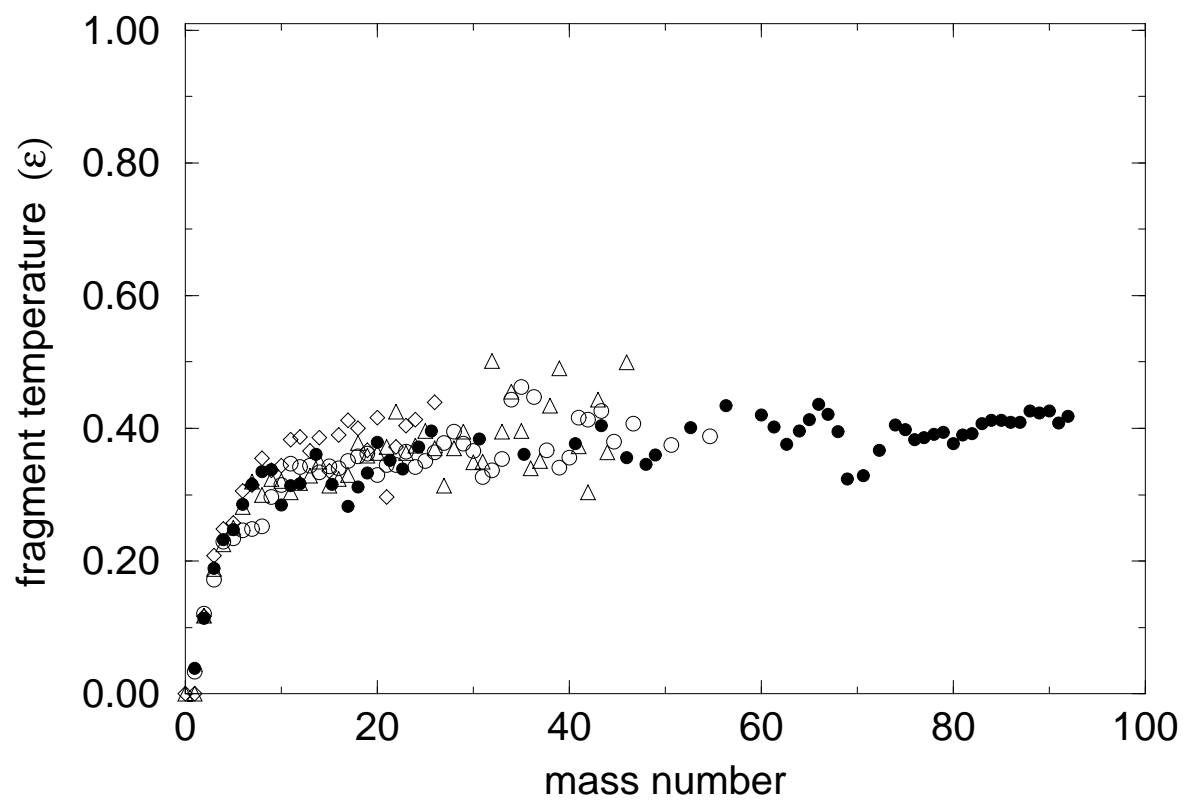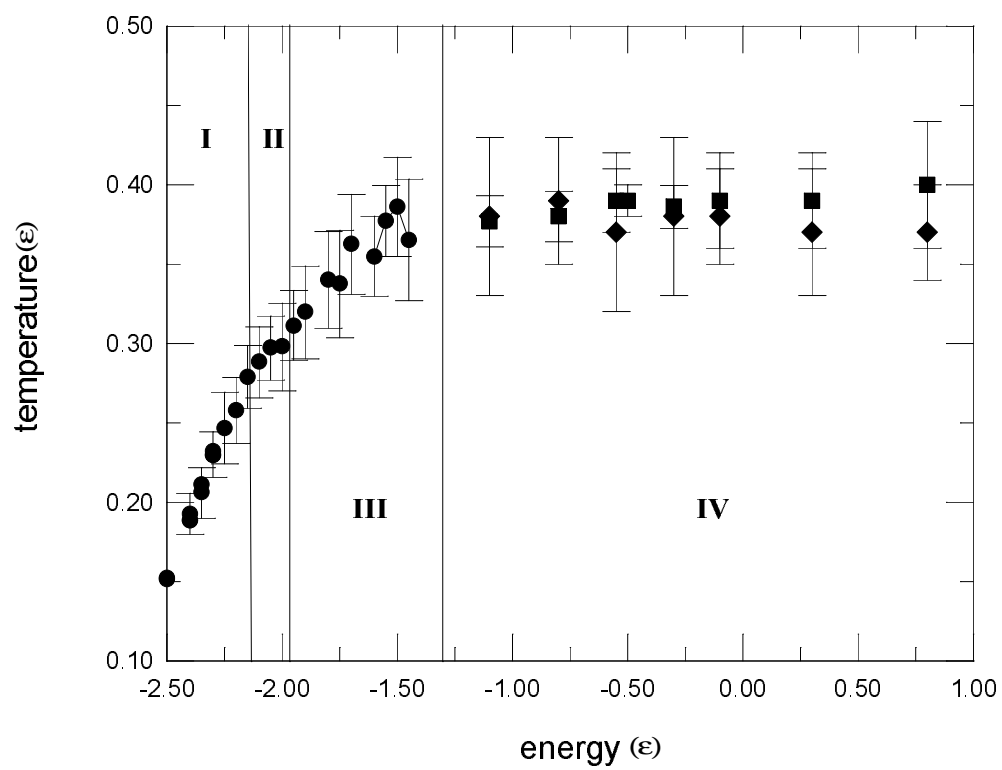
FIG. 1
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FIG. 2

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FIG. 3.
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